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Spontaneous fission properties of ^{252,254}No and ^{256,258}[104] and the disappearance of the outer fission barrier

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Abstract

We have measured the mass and total kinetic energy distributions from the spontaneous fission of ^{252}No, ^{254}No, ^{256}[104] and ^{258}[104]. The results, in combination with earlier measurements for ^{256}No, ^{258}No and ^{262}No, show a sharp transition from asymmetrical mass division in ^{256}No to symmetrical division for ^{258}No and ^{262}No. However, all isotopes of element 104, including ^{260}[104], appear to yield broadly symmetrical mass distributions. The total kinetic energies around 200 MeV for the element 104 isotopes indicate that they fission by the low-energy mode of bimodal fission. Recent calculations of static potential energy surfaces including higher-order asymmetric deformations suggest that the outer fission barrier is well below the ground state in energy, which accounts for the observance of the symmetric mass division.

1. Introduction

In the actinides, the total fission barrier, after shell effects are corrected for, consists of an inner and outer barrier in deformation space. The inner barrier, which arises from the liquid-drop component, is lowest for symmetrical deformations, whereas the outer barrier is lowered by several megaelectron volts for asymmetric deformations compared with symmetric ones [1]. Calculated single-dimension barriers showing the decrease of the outer barrier with increasing Z and N can be seen in Fig. 1. The introduction of shell effects allows mass asymmetry either by a flow of nucleons between emerging fragments that favor closed-shell products [2] or by changing the essential nature of the fission barriers such that odd multipolarity oscillations offer a lower energy path toward scission than even ones [1]. Thus, passage through the outer barrier leaves the nucleus asymmetrically deformed and, presumably, leads to asymmetric mass division. As atomic and neutron numbers increase beyond Pu, the outer barrier decreases in height and may even disappear below the ground state of the heaviest nuclides [3–5]. When this occurs, one might assume that mass distributions would revert to symmetric ones, as they do for fission in the pre-actinide elements. This assumption follows from our knowledge that the inner fission barrier is stiff to asymmetric deformations and minimized by symmetric ones [2].

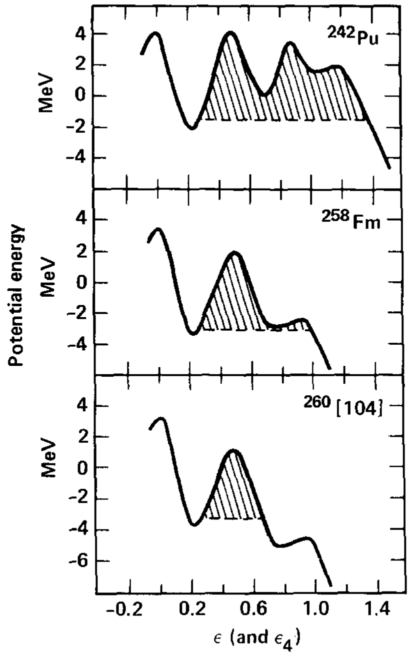

Fig. 1. Fission barriers calculated with single-particle and pairing corrections to the liquid-drop barriers (After Randrup *et al.* [4].)

We had earlier determined the mass and total kinetic energy (TKE) distributions from the spontaneous fission (SF) of the heaviest isotopes of Fm, Md, No and element 104 and had observed two modes of symmetric mass division [6]. One was associated with very high TKEs, around 233 MeV, whereas the other exhibited

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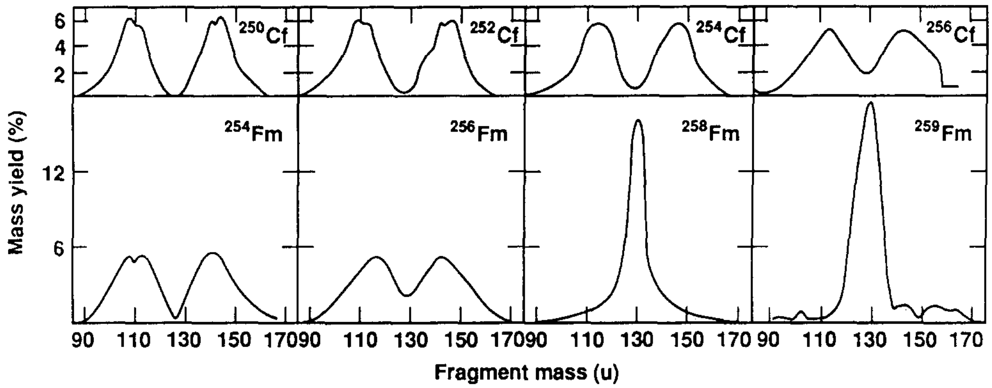

Fig. 2. Mass distributions of fragments obtained in the spontaneous fission of Cf and Fm isotopes. Until ^{258}Fm is reached, only slight differences are found in the asymmetric mass distributions. An abrupt transformation to sharply symmetric mass distributions occurs at ^{258}Fm.

TKEs around 200 MeV. We suggested the latter mode was due to the dropping of the outer barrier below the ground state, in accord with the hypothesis of the last paragraph. As can be seen for the Fm isotopes in Fig. 2, there is a very sharp transition from asymmetric mass distributions to symmetric ones between ^{256}Fm and ^{258}Fm. At one time, theorists estimated from calculations of the potential-energy surfaces (PES) that the outer barrier had disappeared from ^{258}Fm and was well below the ground state for all isotopes of elements with atomic numbers 104 and greater [2, 4]. Today, calculations of the PES have greater refinements, but these have not been applied to many of the known heavy nuclei. We desired to measure the SF properties of the lighter isotopes of No and element 104 and add them to those we have already studied, to test theories that in this region the outer fission barrier disappears completely, resulting in mass-symmetric fission.

2. Experimental details

2.1. ^{252}No (half-life 2.3 s)

Spontaneous-fission decay comprises 27% of the total ^{252}No disintegrations, with the remainder α decay. Earlier but limited measurements of the fragment-energy distributions were made by Bemis *et al.* [7] and Lazarev *et al.* [8] for the SF of ^{252}No. We concluded that it was worth repeating their measurements because we could produce over ten times as many fragment-correlated events and also eliminate any SF background from other reaction products. We produced this isotope by the ^{244}Cm (^{12}C,4n) reaction, and transported the recoil products via an He jet with a KCl aerosol to our MAD (Multiple Alpha Detector) system [9]. The products were deposited by the He jet on 50 μg cm^{-2} films of polypropylene located on rings at the outer edge of a 44.5 cm diameter wheel. A schematic diagram showing the principles of the system is given in Fig. 3. This wheel is rotated in 6° steps at fixed time intervals by a stepping motor so as to center the plastic films between opposing pairs of surface-barrier detectors. With 60 of these plastic films spaced evenly around its circumference, the wheel was indexed to sequential counting stations every 4 s, taking an average of about 0.2 s to do so. Thirty-two detectors above and 32 below the wheel are situated on the same radius as the films and measure the energy deposited by the coincident fission fragments. The first five detector pairs and pairs 27 to 32 were activated for fission counting, while detector pair 6 recorded α spectra that we could later analyze for residual ^{252}No and other reaction products from the bombardment. All fission pulses were digitized by CAMAC analog-to-digital converters and stored together with the clock time of their appearance by our computer. Energy calibrations were carried out with sources of ^{252}Cf electroplated on the same polypropylene films.

We estimate that our efficiency for the He-jet transport of ^{252}No was 35%, a value about equal to other

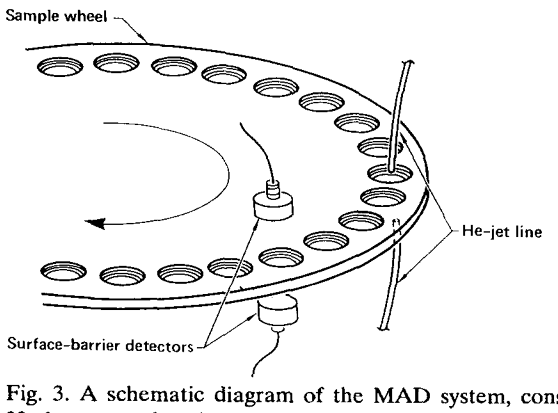

Fig. 3. A schematic diagram of the MAD system, consisting of 32 detectors placed above and below thin plastic films carried by a rotating wheel. Atoms recoiling from the target are carried by an He jet containing a KCl aerosol and are deposited on the surface of the plastic films. The wheel is rotated at fixed time intervals by a stepping motor.

He-jet transport efficiencies we have determined from other ^{12}C-fusion reactions. Overall, we obtained 1741 coincident fission events, which can be compared with 154 events found by Bemis *et al.* [7] and 178 noncoincident fissions found by Lazarev *et al.* [8]. We found a half-life of 2.44 ± 0.12 s for the observed SF activity, in excellent agreement with the total half-life of 2.30 ± 0.22 s obtained by measuring separately the decay of ^{252}No by both the α and SF modes [7]. We observed no long-lived fission activities in the detectors located far from the jet-collection position. In addition, the formation reaction we had chosen for ^{252}No was incapable of producing 2.6 h ^{256}Fm, a common contributor to a longer-lived background.

2.2. ^{254}No (half-life 55 s)

The SF branch for ^{254}No is over 100 times smaller than it is for ^{252}No, making a determination of its fission properties much more difficult. The SF branch has been reported by Türler *et al.* [10] as 0.25% and by Lazarev *et al.* [11] as 0.17%. We produced ^{254}No by two different reactions: ^{246}Cm (^{12}C,4n) at an energy of 72.5 MeV, and ^{245}Cm (^{13}C,4n) reaction at 71.5 MeV, in order to eliminate an SF background caused by a gradual release of the ^{246}Cm target material, which has an SF branch of 0.026%. For these experiments, we also used the MAD system described previously and shown in Fig. 3. In these experiments, the wheel was rotated 6° in 25 s intervals; the first nine detector pairs and pairs 30 and 32 were converted to the measurement of fission-fragment energies. The tenth pair recorded α spectra.

From a series of eight bombardments lasting 180 h, we obtained a total of 287 coincident SF events. Of these, 38 were determined to be due to SF of ^{246}Cm that had been thermally evaporated from the target, carried by the He jet, and deposited on the foils. This number of background events was subtracted from the total mass and TKE distributions by modeling the ^{246}Cm distributions after those of ^{252}Cf.

In bombardments for which the ^{245}Cm (^{13}C,4n) reaction was used to produce ^{254}No, and with no long-lived background detected, we obtained a half-life of 53 ± 20 s. From the number of net SF events and a previously determined transport efficiency of 35% for the He jet, we calculated a cross-section for the ^{246}Cm (^{12}C,4n)^{254}No reaction of 0.4 μb. The SF branch was determined from the ratio of α to SF events to be 0.17 ± 0.02%, in very good agreement with the value from Lazarev *et al.* [11].

2.3. ^{258}[104] (half-life 13 ms)

This nuclide was produced by Ghiorso and coworkers [12] from ^{12,13}C-ion reactions with a ^{249}Cf target. Excitation functions for the formation of this 11 ± 2 ms SF activity were consistent with an assignment to ^{258}[104]. Later, observations of a 13 ± 3 ms SF half-life were made by Somerville *et al.* [13] in bombardments of ^{246}Cm with ^{16}O ions. In addition, researchers using the SHIP velocity filter at the Gesellschaft für Schwerionenforschung (GSI) measured fragment energies from the SF of ^{256}[104] and ^{258}[104] from recoil atoms implanted into the surface-barrier detectors [14]. After calculated corrections for the amount of energy lost by exciting fragments and the pulse-height defect, they arrived at average TKEs of 207 and 220 MeV respectively for these nuclides.

We produced ^{258}[104] from the ^{249}Cf (^{12}C,3n) reaction with 70.2 MeV ^{12}C ions. Because of the shortness of the half-life, we revived the SWAMI apparatus (Spinning Wheel Analyzer for Millisecond Isotopes), which has been described before [6]. A schematic diagram of the SWAMI apparatus is shown in Fig. 4. The wheel can be rotated at preset speeds between 10 and 5000 revolutions per minute (rpm). We used a speed of 463 rpm in these experiments in order to maximize the fraction of the 13 ms activity decaying between the detector pairs.

Calibrations of the detectors for energy and pulse-height defect were made as described previously with 9.5 mm diameter ^{252}Cf sources which had been vacuum-evaporated onto 50 μg cm^{-2} Al foils and overcoated

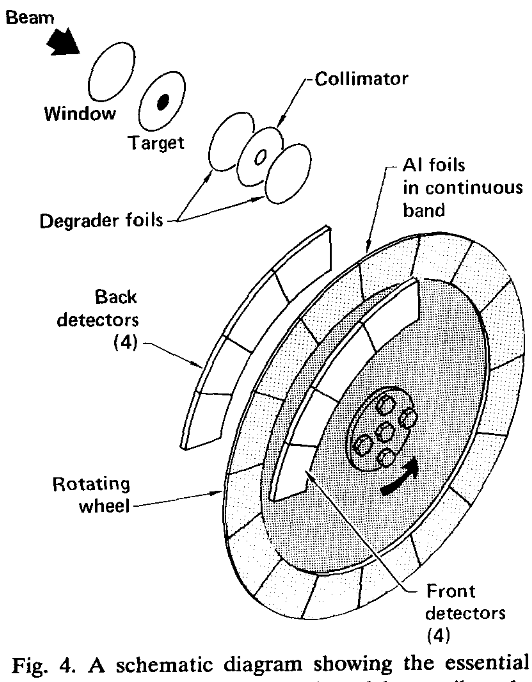

Fig. 4. A schematic diagram showing the essential features of the SWAMI instrument. A portion of the recoil products emerging from the target is stopped in 100 μg cm^{-2} Al foils attached to frames at the edge of a continuously rotating wheel. These foils rotate between pairs of surface-barrier detectors that measure the energy deposited by coincident fission fragments when the product nuclei decay by spontaneous fission.

with an additional approx. 57 μg cm^{-2} of Al to produce a sandwich with a thickness closely approaching our normal 100 μg cm^{-2} Al collector foils on the rotating SWAMI wheel [6].

After 148 h of bombardments, we had collected 274 coincident-fission events. Additional experiments were performed to search for α-emitting nuclides that might also contribute to an SF background. We observed α-energy peaks due to ^{254}No, ^{255}No, ^{250}Fm and ^{252}Fm. However, the amounts collected were 50 to 1000 times less than expected from the excitation functions given by Ghiorso *et al.* [12] and none would have provided an SF background contribution.

Unaccountably, we observed a small number (10) of fissions from counting a foil set with the SWAMI detectors for 80 h after a bombardment. In a separate experiment, we collected recoil products on a single Al foil in a 2 h irradiation and counted fissions for 60 h. Thirteen SF events were detected in the first 22 h and none in the next 38 h. The apparent half-life of this activity was 8^{+4}_{-2} h. We calculated that this SF activity produced 13 fission events, and subtracted this amount from the gross mass and TKE distributions after assuming that its mass and TKE distributions were like those of ^{252}Cf. The question arose as to the source of this SF activity because none is obvious with this half-life within the range of nuclides that can be produced by reactions of ^{12}C with ^{249}Cf. We suggest that an electron-capture-delayed fission isomer, arising from 8.6 h ^{250}Es, is the most likely possibility. A similar beta-delayed fission isomer with this branching ratio has recently been discovered in 7.6 h ^{256m}Es [15].

We derived a half-life of 14 ± 2 ms, which agrees with all previous measurements. We obtained a formation cross-section of 8 ± 5 nb, which is two to three times less than we anticipated, in as much as Bemis [16] had found this cross-section to be about 20 nb and the JORPLE program predicted 26 nb [17]. However, our cross-section for the 3n reaction is consistent with the 10 nb reported for the ^{249}Cf (^{12}C,4n) reaction [12].

2.4. ^{256}[104] (half-life 6 ms)

First reported in 1975 as a 4.8 ms SF activity produced in the reaction of ^{208}Pb with ^{50}Ti ions [18], this identification was later confirmed at GSI using the same nuclear reaction and by separating the recoils from the ion beam with the velocity filter SHIP [19]. The latter group obtained a half-life of 7.4 ms and, as noted under the section on ^{258}[104], they sought to determine the TKE. We produced a 6.6 ± 1.1 ms SF activity from bombardments of ^{244}Cm with 93.8 MeV ^{16}O ions. The cross-section estimate by the JORPLE code for this reaction was 3.4 nb. Again the SWAMI apparatus, with the wheel rotating at 740 rpm, was employed to measure the energies of coincident fission fragments. We continued the fission counting of a group of foils after the end of their irradiation, and observed only a single SF event, indicating no appreciable background. In addition, we collected recoil products on a single foil during a 30 min bombardment, which was then analyzed by α counting to show that ^{256}Fm may contribute, at most, 3% of the observed fissions. The agreement between our 6.6 ms half-life and the accepted value of 7.4 ms argues against appreciable fissions from any source other than ^{256}[104].

After 160 h of operation time at the cyclotron, we had accumulated only 28 SF events, or about one every 2 h of actual fission-counting time. Continuation of these experiments to obtain several hundred events for statistical significance was futile at this rate, and we abandoned further work. Furthermore, many of the special trapezoidal detectors were affected by radiation damage and were no longer stable with respect to large losses in fission-signal amplitude during the bombardments. Though we would have preferred to observe ten times this number of events, our mass and TKE distributions contain much of the information we desired.

3. Results and conclusions

Our mass and TKE distributions determined for the SF of ^{252}No and ^{254}No are shown in conjunction with those of ^{256}No [20], ^{258}No [6] and ^{262}No [21] in Fig. 5. Their mass distributions are asymmetric with a shallow valley between the mass peaks, not unlike those of ^{254}Fm and ^{256}Fm. Our average post-neutron TKE of 194.3 MeV for ^{252}No and 189.2 MeV for ^{254}No are also comparable to the values of 193.1 MeV for ^{254}Fm and 195.9 MeV for ^{256}Fm, although the value for ^{254}No seems somewhat low. Even though the TKE obtained for ^{252}No by Bemis *et al.* [7] in their earlier study is larger than ours by some 8 MeV, all but about 2 MeV of this difference can be accounted for by their correcting for neutron evaporation to a preneutron TKE and by their use of a TKE of 183.1 MeV for ^{252}Cf in the energy calibrations of their detectors rather than the most recent parameters and a value of 181.0 MeV that we used [22]. Generally, our results for the SF properties of ^{252}No confirm the main features found in their investigation.

When the distributions for all No isotopes are considered, there is little to distinguish the trends from those found for Fm isotopes. As N increases, the mass distributions evolve from asymmetric to broadly symmetric to sharply symmetric. In the same evolution, the TKE distributions hardly change until ^{258}No is

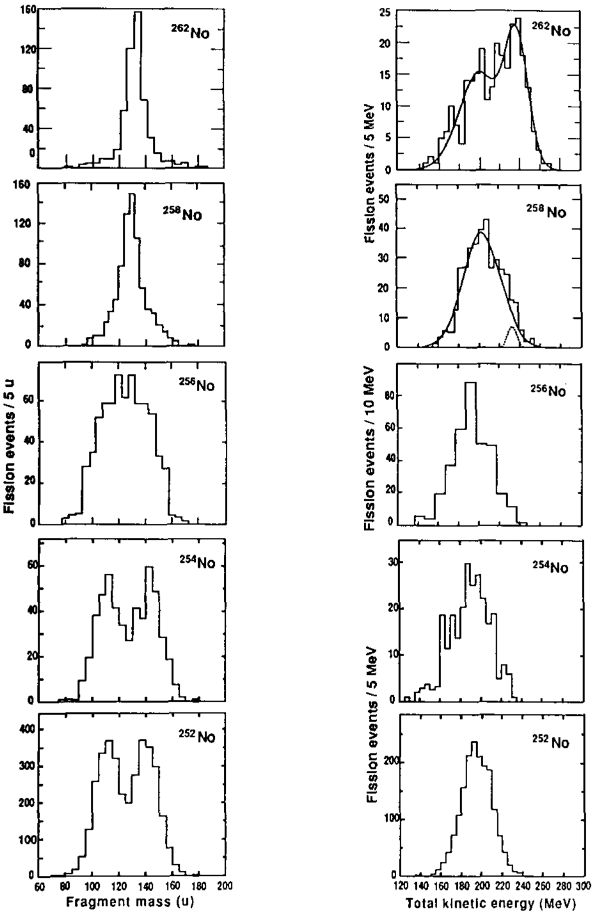

Fig. 5. Provisional mass and total kinetic energy distributions measured for the isotopes of No. The distributions for ^{256}No were determined by Hoffman *et al.* [20], ^{258}No and ^{262}No are from Refs. 6 and 21 respectively and the remainder are reported here. Note the sharp change in mass distributions from asymmetric to symmetric as N increases. Similarly, the TKE distributions shift to predominantly the high-energy mode.

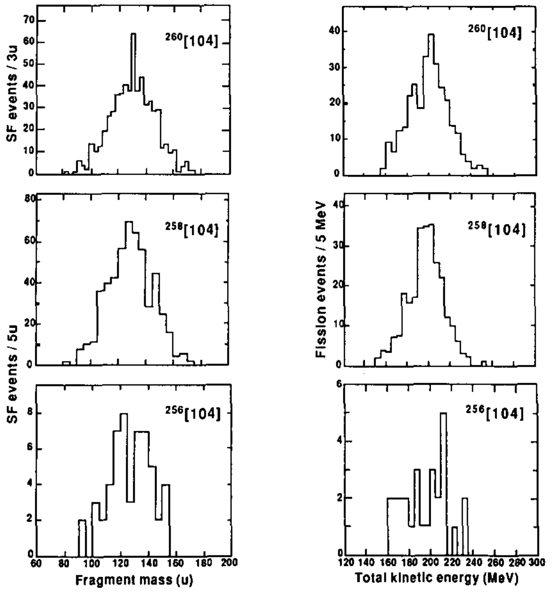

Fig. 6. Provisional mass and total kinetic energy distributions from spontaneous fission of the isotopes of element 104. Those for ^{260}[104] are from previously reported work [6]. Note that mass symmetry and TKEs around 190–200 MeV tend to prevail throughout.

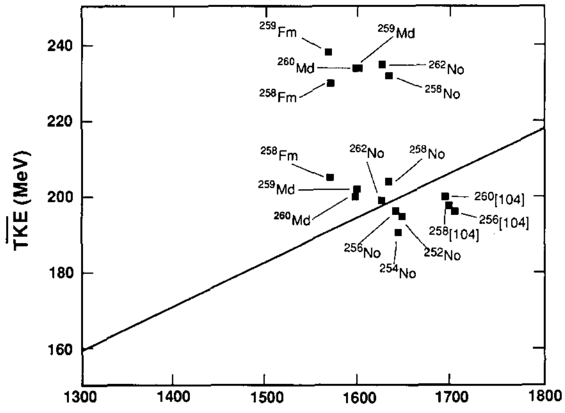

Fig. 7. Comparison of expected liquid-drop TKEs (——) with the TKEs obtained for nuclides reported here and for the two groups of TKEs found for the bimodal nuclides. The line is defined by the best fit of experimental TKEs to a linear dependence on the Coulomb parameter, $Z^2/A^{1/3}$, in the liquid-drop model of fission.

reached and the first hint of a high-energy component becomes evident. With ^{262}No, the high-energy mode of bimodal fission predominates and the mass distribution becomes remarkably narrow. These characteristics are the same as we had noted earlier for the Fm and Md isotopes in that these transitions, when they do occur, are very abrupt, within one or two neutrons [6].

Figure 6 illustrates the SF properties of the isotopes of element 104 that we have measured, including those of ^{260}[104] reported earlier. None appears to have bimodal properties, distinguished by a second TKE distribution peaking around 233 MeV. There may be some hint of this mode in ^{260}[104], where a small number of events are seen clustering close to the Q value of 256 MeV for this fission reaction. The average TKE for ^{256}[104] is 198.9 ± 4.4 MeV and for ^{258}[104] it is 197.6 ± 1.1 MeV, both very close to the value of 200 MeV that characterizes the low-energy mode found in six of the heaviest nuclides studied [6, 23]. The mass distributions appear broadly symmetric for all of the 104 isotopes, which appear to follow a fission process that could be described by the liquid-drop model: symmetric mass distributions and average TKEs that fit an empirical extrapolation, shown in Fig. 7 as a line obtained by Viola *et al.* [24].

In order to make substantial conclusions about the role of the outer fission barrier upon mass division for the low-energy fission mode, it seems necessary to know the potential energy difference between the height of this barrier and the ground state. In principle, this energy could be found from macroscopic–microscopic calculations that minimize the static potential-energy surface through all possible deformations. However, such detailed calculations have not yet been made for many of the nuclides we have measured. Another source of such energy comes from phenomenological studies relating SF half-lives to the first and second barrier heights [25]. In this work, half-lives and energies for the isomeric fission decay of Th, U, Pu, Cm and Cf isotopes were used to normalize relative barrier heights for heavier nuclides. The energies they derive for the heights of the outer barrier for the isotopes of No and element 104 range from 2.3 to near 0 MeV, all decreasing sharply for nuclei beyond the 152-neutron subshell. These values are about 0.9 MeV larger than those from the calculations of Möller and Nix [26] performed in 1981 and are larger, still, than those given by recent PES calculations noted below.

When compared with heights of outer barriers from early PES calculations, our findings on the mass distributions as Z and N change bear a close resemblance to predictions made many years ago concerning where the outer fission barrier dropped below the ground state. Randrup *et al.* [4] reported that their calculations showed that this condition was reached in the Fm isotopes at mass 260 and existed for all of the isotopes of element 104. This was offered as an explanation for the unexpected change between Fm and [104] in empirical trends of SF half-lives because, for the latter nuclei, only a single rather than a double barrier had to be penetrated. Mustafa and Ferguson, using the two-center shell model, had outlined regions where asymmetric and symmetric mass division would prevail [2]. However, their estimates for the mass-symmetric region were based on the possibility of the fissioning species evolving into two spherical fragments at or near closed nucleon shells, which characterizes our high-energy mode. They had not yet recognized the possibility of a low-energy mode that would yield symmetric mass distributions, but they did note that the outer fission barrier disappeared for all nuclei with $Z \geqslant 106$. For comparison with our data, these earlier calculations of the PES using the Strutinsky–Nilsson method are limited because they did not include all the isotopes we have studied, they were often restricted to quadrupole deformations, and the existence of new valleys after the first barrier was unknown at the time.

Newer calculations of the PES now include hexadecapole deformation and some include odd-multipolarity vibrations to as high as $\ell = 7(\beta_\ell)$ [27]. The inclusion of higher-order asymmetric shapes is important because it was found for ^{258}Fm, as is shown in Fig. 8, that the outer barrier leading to deformed shapes virtually disappears and is about 2 MeV below the ground state when these higher orders of multipolarity are considered [27, 28].

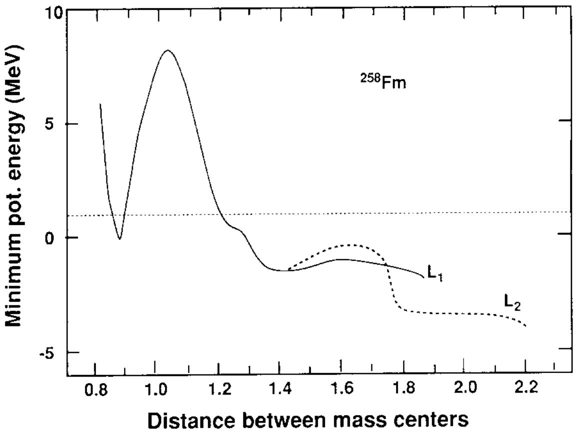

Fig. 8. Fission barriers of ^{258}Fm along compact (L_1) and elongated (L_2) shapes. Both symmetric and asymmetric deformations from $\ell = 3$ to 6 were considered in calculating the potential-energy surface [27]. Of importance is that the residual outer barriers in both valleys are below the ground-state energy shown by the horizontal dotted line.

If limited to odd multipolarities with $\ell \leqslant 3$, Möller and colleagues create an outer barrier for ^{258}Fm that is still 0.15 MeV above the ground state [29]. Thus, it seems that massive PES calculations allowing a wide range of shape degrees of freedom and encompassing all of the nuclides we have studied would enable us to offer more than a qualitative comparison with our experimental results. Nevertheless, we should note that even now, the most detailed barriers minimized on the potential-energy surfaces support our notion that if there is no outer barrier to penetrate, SF produces mass-symmetric division.

Acknowledgments

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